METAL VAPOR VISIBLE LASER KINETICS PROGRAM

FINAL TECHNICAL REPORT

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4. Electronically Excited Bismuth Atoms
5. Metastable Atom Quenching
6. Resonance Absorption
7. Resonance Fluorescence
8. Electron Excitation Cross Sections
9. Electron Mixing Cross Sections

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Objective: The objective of this program is to investigate experimentally and theoretically the important kinetic rate processes pertinent to the development of a high power metal vapor visible laser. This research consists of three tasks. Each of the tasks are summarized below.

TASK I - LOWER LEVEL KINETICS (EXPERIMENTAL)

Objective: The purpose of this task is to identify metal atom systems that allow selective collisional relaxation processes to efficiently quench lower

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levels of potential or demonstrated laser transitions. The present experimental study is directed to kinetic rate constant measurements for collisional relaxation of low-lying metastable states of lead, copper, and bismuth.

Accomplishments: The high temperature flash photolysis apparatus utilizing resonance absorption detection techniques developed under this contract was used to provide temperature sensitive collisional quenching rate constants for lead, copper and bismuth. These subtasks have been completed and were reported in various semi-annual technical reports as follows:

lead($6p^2$) $^3P_2$ and $^3P_1$ states	- Semi-annual Technical Report March 1975
copper($3d^9 4s^2$) $^2D_{5/2}$ state	- Semi-annual Technical Report February 1976
bismuth($6p^3$) $^2D^o_{3/2}$ and $^2D^o_{5/2}$ states	- Semi-annual Technical Report April 1976
	- This Report

TASK II - UPPER LEVEL KINETICS (EXPERIMENTAL)

Objective: The purpose of this task is to determine the efficiency of various quenching gases in deactivating the upper laser level of lead. To be useful, a quenching gas must rapidly relax the lower laser level and slowly relax the upper laser level, i. e. be selective.

Accomplishments: The high temperature resonance fluorescence apparatus utilizing DC phase sensitive detection techniques constructed under this contract was used to provide cross-section information on the lead(6p7s) $^3P_1^o$ state. This task was completed and results reported in the semi-annual technical report dated February 1976.

TASK III - EXCITATION PROCESSES (THEORETICAL)

Objective: The objective of this effort is to calculate electronic impact and quenching cross sections for the upper and lower laser levels of copper.

Accomplishments: Important cross sections required for modeling the copper vapor laser were calculated. These results, when applied to modeling the laser's performance, predict that a large number of atoms are capable of being utilized for lasing, with a correspondingly higher laser energy density and a greater overall system efficiency. The task was completed and results reported in the semi-annual technical report dated February 1976.

FOREWORD

ARPA Order Number: 1806

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Short Title of Work: Metal Vapor Visible Laser Kinetics

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I. INTRODUCTION

In an effort to gain some understanding of the mechanisms whereby electronically excited metal atoms are relaxed in collisions with simple atoms and molecules, the quenching kinetics for all the members of a group have been systematically studied[1-3] (e.g., N, P, As, Sb, Bi). In this way similarities between electronic structure can be utilized in the interpretation of the available rate data. This interpretation is usually discussed within the context of correlation diagrams which are constructed through consideration of energy and symmetry constraints. For the lighter members of the group, on utilizes weak spin orbit coupling approximations[4], whereas for the heavier elements, (J, Ω) coupling seems more appropriate.[5] Often, there has been shown to be a correlation between these symmetry constraints and available rate data. In some cases, however, where no energy accessible correlations exist between reactants and product channels, the rate data still indicate efficient quenching is occurring which implies a process involving non-adiabatic transitions. Examples include possible surface hopping near curve mergings at close internuclear separations[6] and E→V energy transfer processes for collisions involving molecular quenching gases.[7] Choosing between likely relaxation mechanisms is largely speculative since the disappearance of the metastable electronic state of the excited atom is the usual diagnostic. Information on the product channels is scarce.

We report here temperature dependent rate data for the collisional relaxation of the $6p^3\,(^2D^o_{3/2})$ and $6p^3(^2D^o_{5/2})$ states of atomic bismuth.

Temperature dependent rate data naturally provide additional information to consider in the selection of any likely relaxation mechanism.[8] These data were obtained using a temperature controlled, flash photolysis, kinetic spectroscopy apparatus which has been used to study temperature sensitive rate data[8,9] for lead $6p^2$ ($^3P_2$) and $6p^2$ ($^3P_1$) and for copper $3d^9\ 4s^2$ ($^2D_{5/2}$) states. Rate constant measurements were made for quenching by Ar, Xe, N_2, H_2, D_2, CO, O_2, CO_2 and SF_6 at 300, 450 and 550°K. The data are reduced to provide Arrhenius parameters where appropriate and likely relaxation mechanisms are discussed.

II. EXPERIMENTAL

The general technique and experimental approach has been described in earlier publications.[3,6,8,9] The apparatus is shown schematically in Fig. 1. It consists of a Glomax hollow cathode light source, a 45 cm long reaction cell (1.8 cm ID) constructed of suprasil quartz with quartz windows fused to the cell itself, and a mechanical shutter/monochromator/photomultiplier detection system[10] for monitoring the resonance line chosen for the time resolved absorption measurements. The reaction cell is heated resistively by passing current through Nichrome heating wire which is wound in a spiral over the entire surface of the cell at approximately 8 mm intervals. In the cell, temperatures in excess of 600°K were easily obtained as determined by thermocouples (Iron-Constantan) placed at various locations on the glassware surface. This high temperature capability allowed us to very effectively degas the photolysis cell (residual pressures of 10^{-5} torr and leak plus outgassing rate of less than 0.1μ/min).

The overall procedure was the same as that used in the previous room temperature experiments.[3] Premixed gases were introduced into the evacuated reaction chamber. Nonequilibrium concentrations of the $^2D^o_{5/2}$ and $^2D^o_{3/2}$ of bismuth were generated by the flash photolysis of a trace (approximately 3 ppm) amount of trimethyl bismuth (TMB) in the presence of argon buffer gas and added quenchers. The decay of the metastable $^2D^o_{5/2}$ and $^2D^o_{3/2}$ states was monitored by time resolved

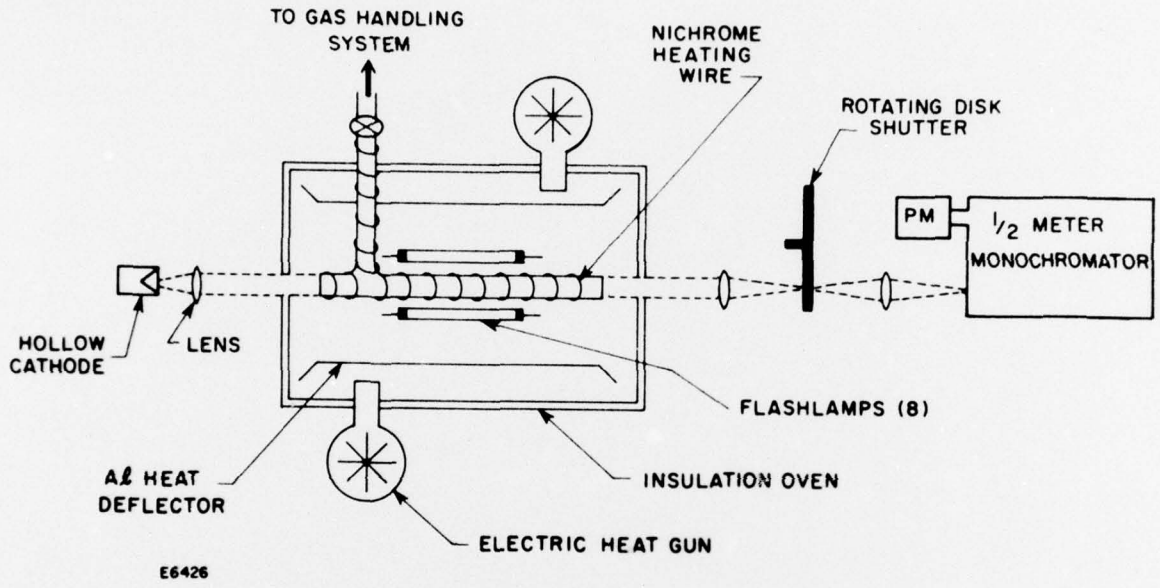

Fig. 1 Diagram of the Apparatus

line absorption techniques at 2938 and 2898Å respectively, beginning at times typically 100μ sec after the photolysis flash. The usual modified Beer-Lambert Law was used as discussed in our earlier publications.[3] The system was baked under vacuum at a temperature higher than the highest one used for kinetic measurements. Rate constants were routinely checked at room temperature to establish that the results were in agreement with our previous work.[3] The system was then heated, premixed gases admitted to the hot cell and measurements made at temperatures up to 550°K.

Cylinder grade gases were used directly and were of the following purity based on the manufacturer's quoted values: Argon (Liquid Carbonic, 99.998%), Xenon (Research Grade, Matheson, 99.999%), Nitrogen (Liquid Carbonic, 99.996%), Oxygen (Liquid Carbonic, 99.7%), Hydrogen (Matheson, 99.95%), Deuterium (Matheson, 99.5%), Carbon Monoxide (Research grade, Matheson, 99.99%), Carbon Dioxide (Research Grade, Matheson 99.995%), and Sulfur Hexafluoride (Mass Oxygen Equipment Co., 99.8%). The trimethyl bismuth (ALFA Products, 99.99%) was outgassed by several freeze, pump, thaw cycles before being mixed by successive dilutions with argon buffer gas. Quenching rates of bismuth by TMB quoted below are, of course, weighted averages for Bi, unphotolyzed TMB, methyl groups, fragments, etc.

III. RESULTS

Figure 2 shows some typical data for the collisional relaxation of the Bi ($^2D^o_{5/2}$) state in mixes with and without added hydrogen. The slopes of these first order decay plots provide values of $\gamma k'$, where γ is an empirically determined constant and part of the modified Lambert-Beer expression and k' is the pseudo first order rate constant describing all occurring relaxation processes, i.e.,

$$\gamma k' = k_{Ar} [Ar] + \sum_i k_i [N_i] + k_q [Q]$$

where $k_{Ar} [Ar] + \sum_i k_i [N_i]$ represents the decay of Bi due to collisions with the species present in the baseline mix and is taken to be constant. For this example, Q is molecular hydrogen and, therefore, a plot of $\gamma k'$ vs $[H_2]$ provides a relationship for determing the absolute second order quenching rate constant, k_{H_2} (see Fig. 3). Other experiments carried out at 450 and 550°K provide temperature dependent rate data (see Figs. 4 and 5). Similar data were collected for relaxation of both metastable states in collisions with Ar, Xe, N_2, H_2, D_2, CO, CO_2, O_2 and SF_6. These data are summarized in Table I. Where uncertainties are indicated, they represent one standard deviation of a least squares computer fit to the data.

Rate constants are reported as upper bounds when the ratio of the derived rate constant to the gas kinetic rate constant ($k_{GK} = 2 \times 10^{-10}$ cm^3/molecule sec) is less than the mole fraction of manufacturer's stated level

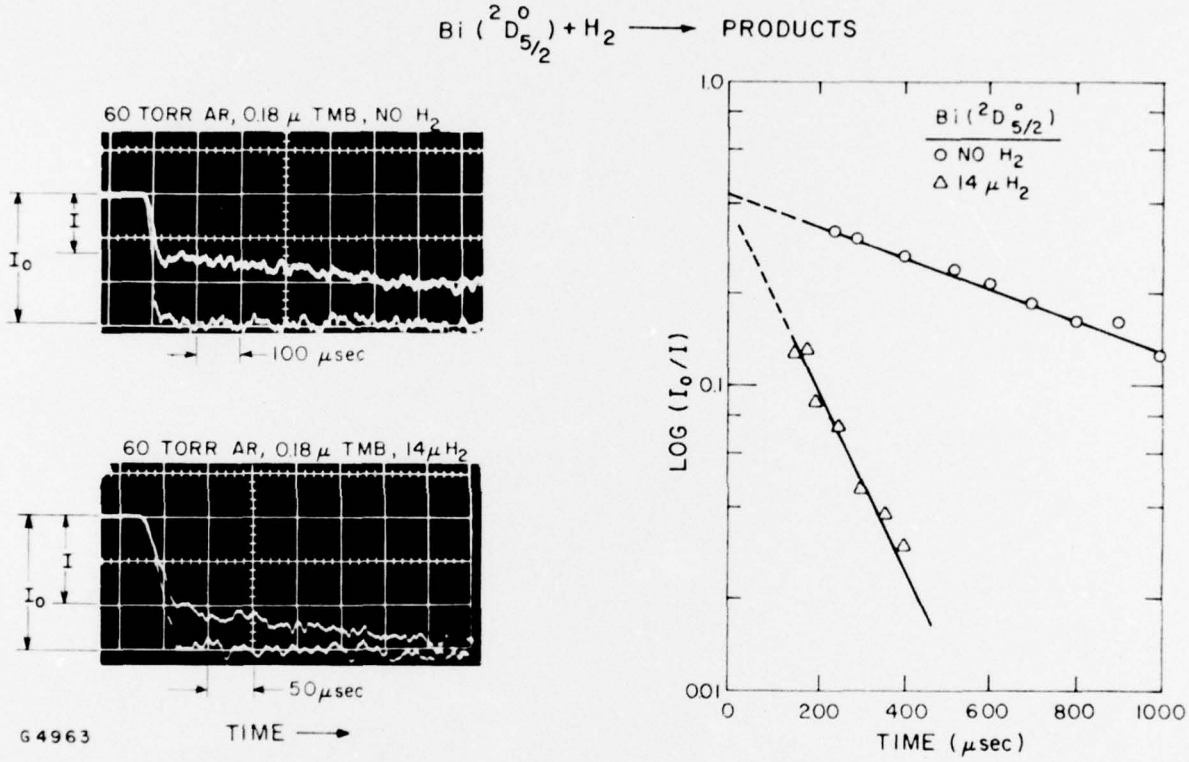

Fig. 2 Collisional Quenching Data for Bi ($^2D^o_{5/2}$) Being Relaxed by Molecular Hydrogen

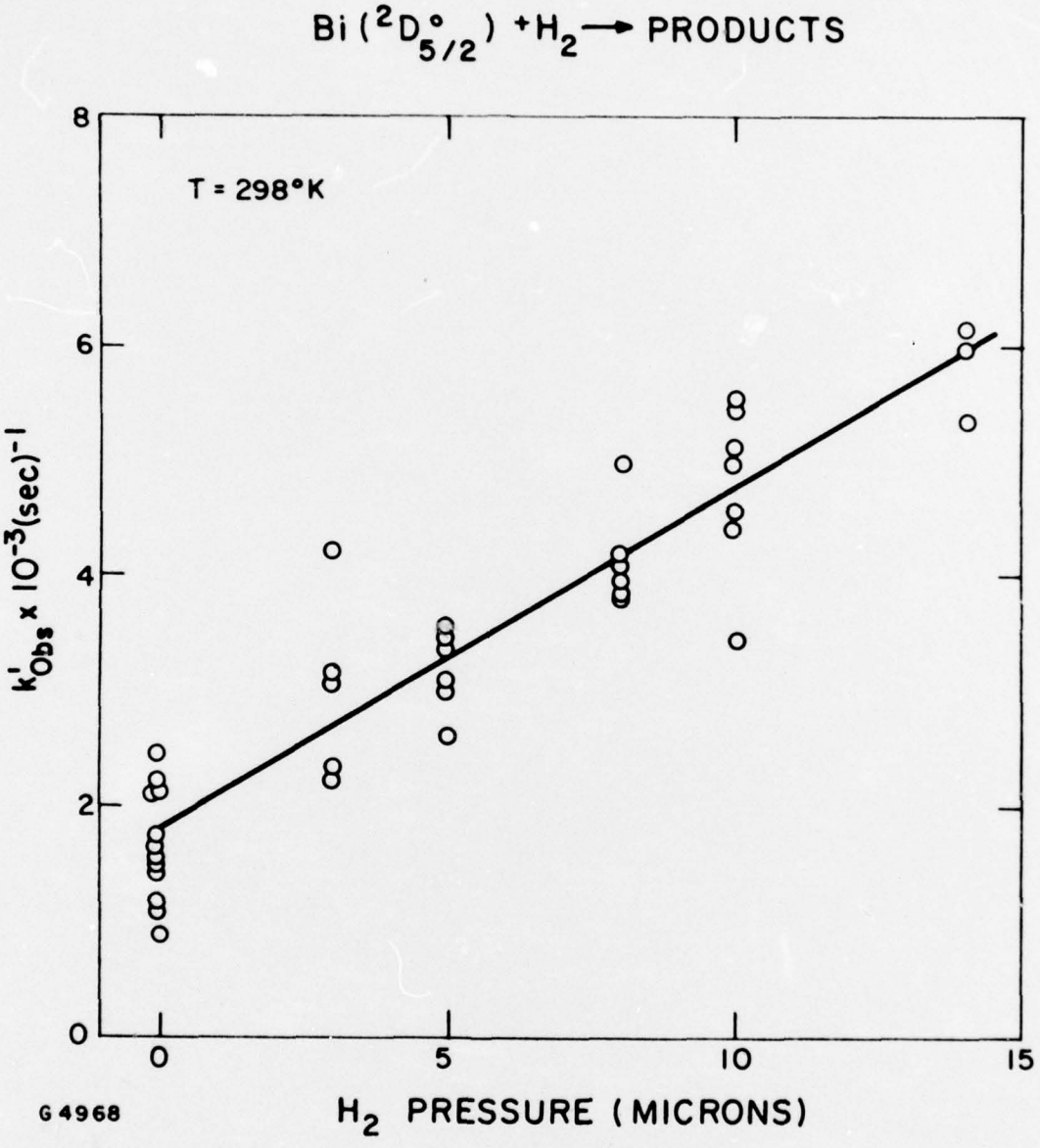

Fig. 3 Plot of Pseudo-First-Order Rate Constant, k'_{obs}, vs P_{H_2} at 298°K

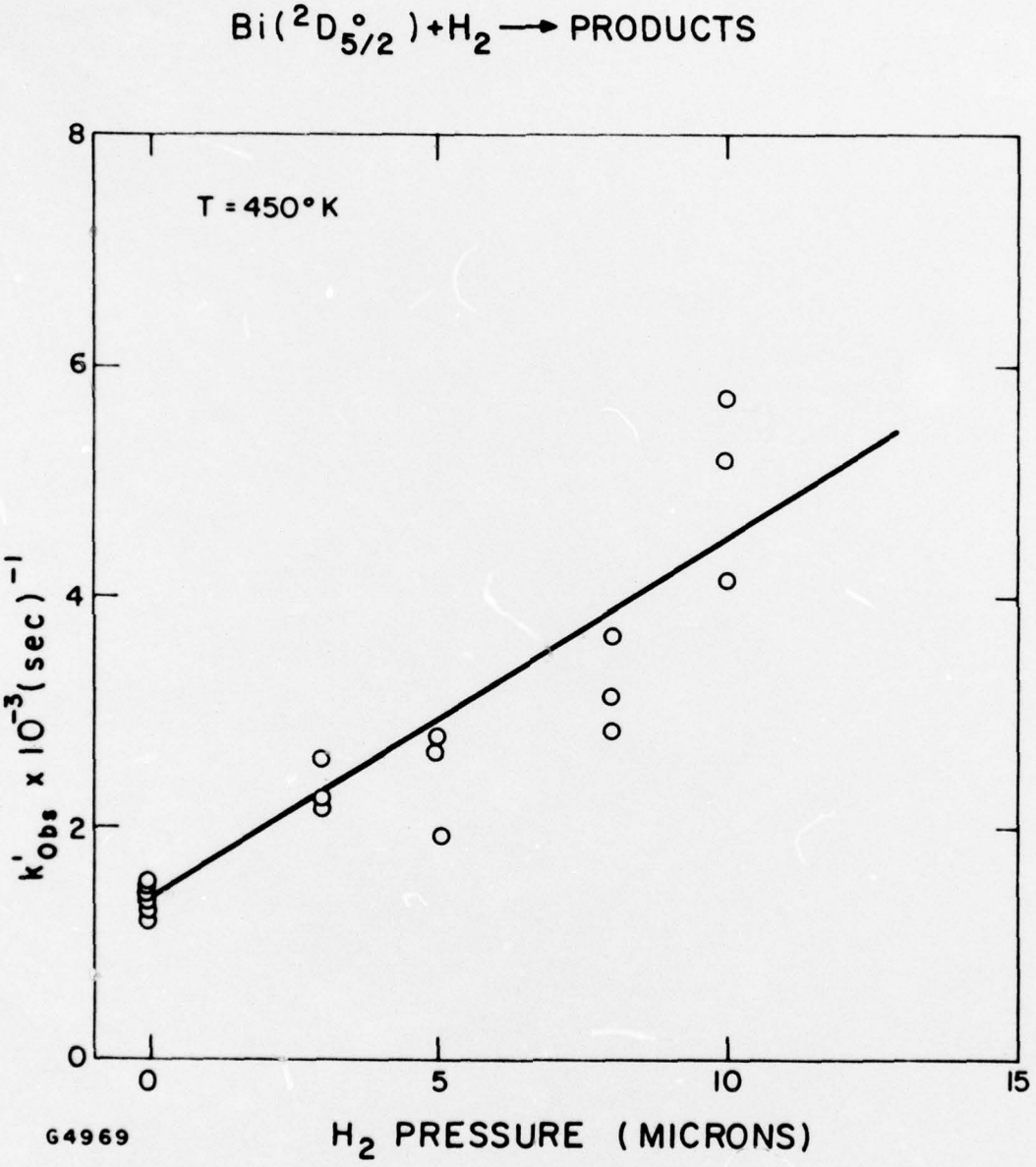

Fig. 4 Plot of Pseudo-First-Order Rate Constant, k'_{obs}, vs P_{H_2} at 450°K

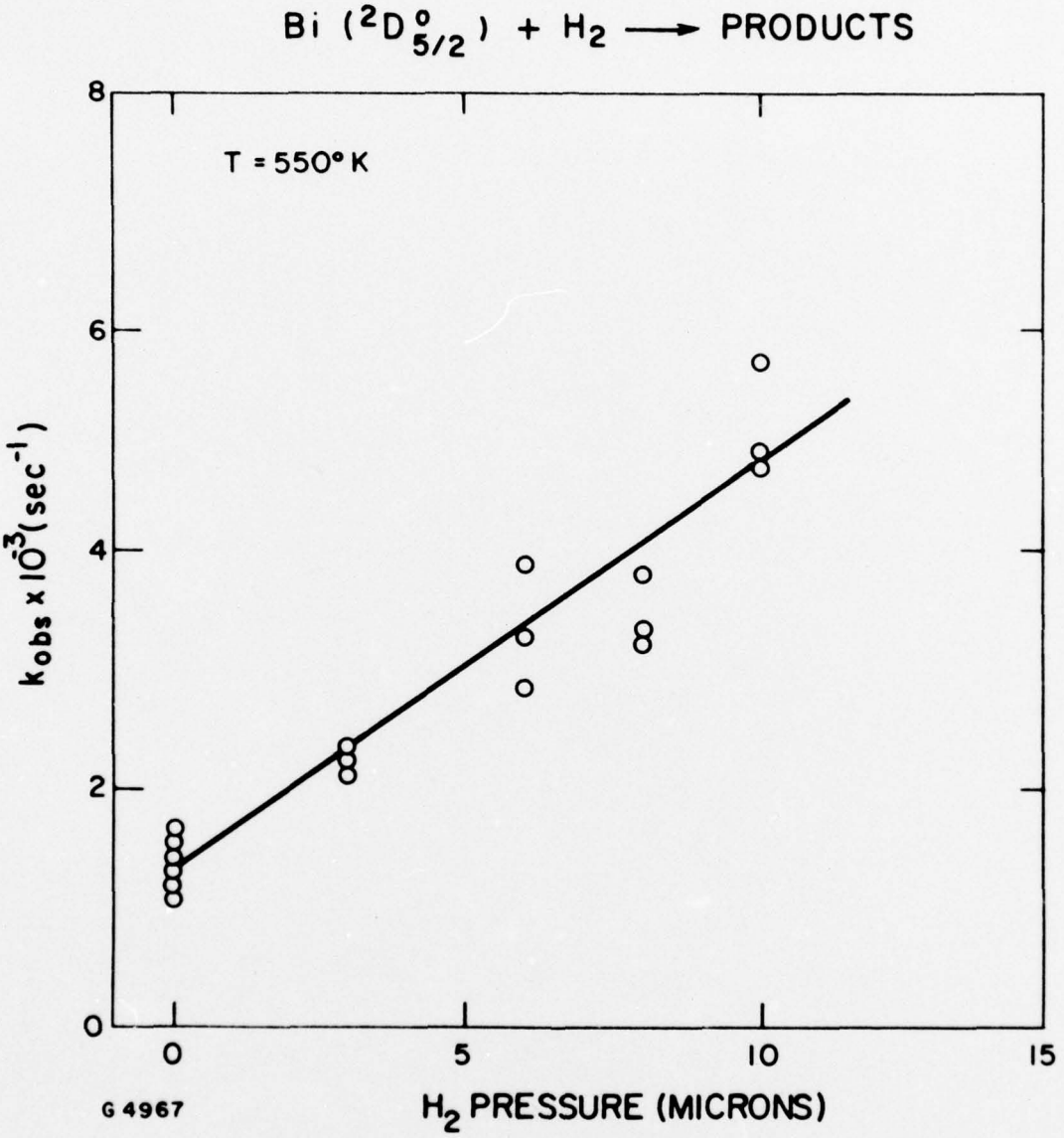

Fig. 5 Plot of Pseudo-First-Order Rate Constant, k'_{obs}, vs P_{H_2} at 550°K

TABLE I

SUMMARY OF KINETIC RATE CONSTANTS AS f (TEMPERATURE)

k (cm^3/sec)

Q	$Bi(^2D_{3/2}^o) + Q \rightarrow$ Products (Temperature, °K)			$Bi(^2D_{5/2}^o) + Q \rightarrow$ Products (Temperature, °K)		
	300	450	550	300	450	550
Ar		no measurable effect			no measurable effect	
Xe		no measurable effect			no measurable effect	
N_2	$<0.0^{+1.9}$ (-16)	$<3.4 \pm 2.3$ (-16)	1.4 ± 0.3 (-15)	$<6.2 \pm 2.9$ (-16)	9.7 ± 0.9 (-15)	2.8 ± 0.1 (-14)
H_2	7.3 ± 0.3 (-15)	1.9 ± 0.1 (-14)	2.9 ± 0.1 (-14)	1.0 ± 0.1 (-11)	1.6 ± 0.2 (-11)	2.2 ± 0.2 (-11)
D_2	$<2.5 \pm 1.6$ (-16)	$<1.4 \pm 0.3$ (-15)	$<9.4 \pm 0.4$ (-16)	1.0 ± 0.1 (-13)	7.3 ± 0.7 (-13)	1.0 ± 0.1 (-11)
CO	$<1.4 \pm 0.6$ (-15)	3.4 ± 0.2 (-14)	1.3 ± 0.1 (-13)	4.7 ± 0.2 (-13)	1.1 ± 0.1 (-12)	1.8 ± 0.1 (-12)
O_2	3.7 ± 0.2 (-13)	1.5 ± 0.1 (-12)	3.7 ± 0.5 (-12)	2.3 ± 0.3 (-11)	2.4 ± 0.2 (-11)	2.5 ± 0.5 (-11)
CO_2	$<1.0 \pm 0.2$ (-15)	1.5 ± 0.2 (-15)	3.0 ± 0.3 (-15)	2.4 ± 0.4 (-14)	4.5 ± 0.3 (-14)	9.2 ± 0.8 (-14)
SF_6	$<2.2^{+3.1}_{-2.2}$ (-16)	$<1.2 \pm 1.0$ (-15)	$<7.8^{+8.3}_{-7.8}$ (-16)	$<3.0 \pm 0.7$ (-15)	$<3.3 \pm 0.7$ (-15)	$<3.4 \pm 1.2$ (-15)

of impurities in the quenching gas. This is necessary because of our uncertainty in assigning the observed increase in $\gamma k'$ to the quencher under investigation or to the impurities which come with the quencher.

These rate data, where appropriate, are plotted in Arrhenius form in Figs. 6 and 7. Table II summarizes the Arrhenius parameters for those cases where the results are not reported as bounds. The error limits for these parameters were estimated from the maximum and minimum values for the slopes and intercepts which could be drawn and be consistent with the data.

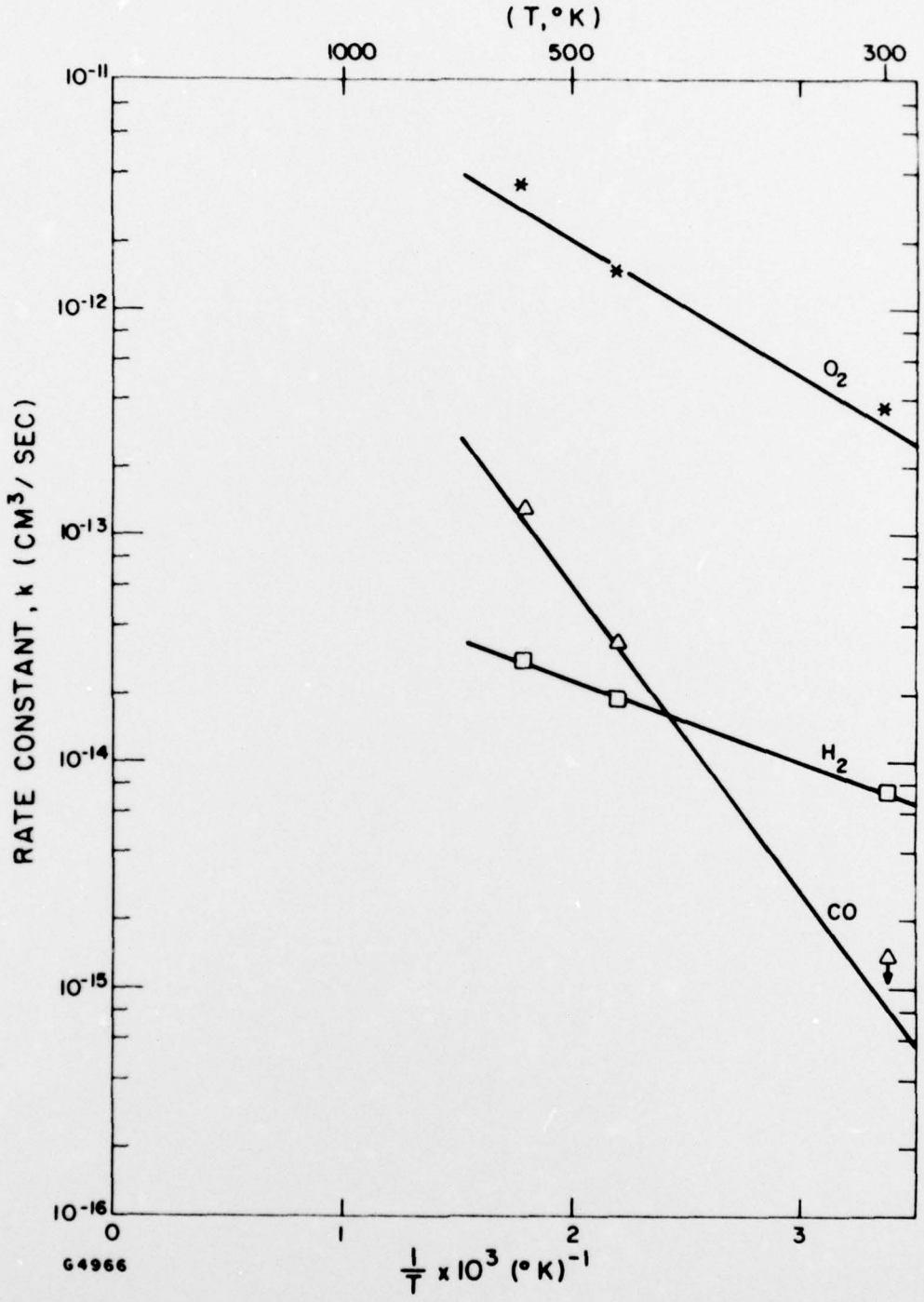

Fig. 6 Arrhenius Plots of k_q (T) for the Relaxation of Bi ($^2D^o_{3/2}$) in Collisions with Q = O_2, H_2 and CO

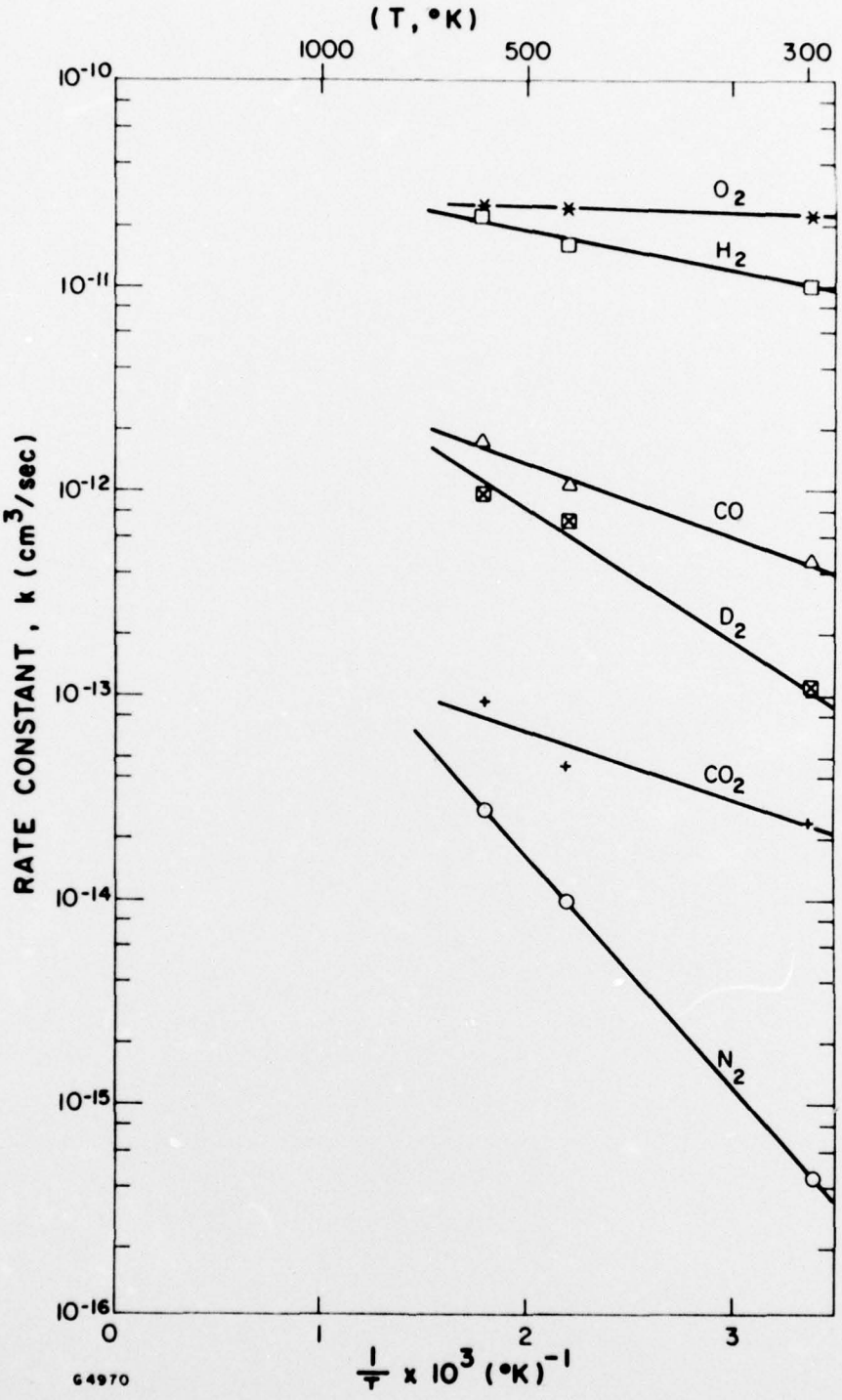

Fig. 7 Arrhenius Plots of k_q (T) for the Relaxation of Bi ($^2D^o_{5/2}$) in Collisions with Q = O_2, H_2, CO, D_2, CO_2 and N_2

TABLE II

KINETIC RATE CONSTANTS

(Arrhenius Parameters)

Reaction	A (cm^3/sec)	E_A (kcal/mole)
Bi ($^2D_{3/2}^o$) + CO →	2.4 ± 0.2 (-11)	5.9 ± 0.4
+ H_2 →	1.3 ± 0.3 (-13)	1.7 ± 0.1
+ O_2 →	4.2 ± 1.5 (-11)	2.9 ± 0.3
Bi ($^2D_{5/2}^o$) + N_2	3.0 ± 2.0 (-12)	5.2 ± 0.5
+ CO_2	3.1 ± 1.5 (-13)	1.5 ± 0.2
+ D_2	1.6 ± 0.6 (-11)	2.9 ± 0.3
+ CO	7.2 ± 3.3 (-12)	1.6 ± 0.3
+ H_2	4.7 ± 1.6 (-11)	0.9 ± 0.1
+ O_2	2.7 ± 1.4 (-11)	0.1 ± 0.3

IV. DISCUSSION

An important aspect of having temperature dependent rate constant information involves the likelihood of choosing an appropriate mechanism whereby the energy represented by the excited metastable states is removed in undergoing an effective relaxation collision. From the data listed in Table II and summarized in Figs. 6 and 7, discussion of these results is limited to H_2, CO, and O_2 for both metastable states and N_2, D_2 and CO_2 for the Bi $^2D^o_{5/2}$ state.

A starting point for discussion is to consider the validity or appropriateness of the (J, Ω) correlation diagrams.[5] This approach certainly has limitations[1] but should serve as an improvement over simple energy and spin considerations. Many of the diagrams for these metastable states are conveniently available as reported in the room temperature work of Bevan and Husain.[2] For example, the reaction

$$Bi\ (^2D^o_{3/2}) + CO_2\ (^1\Sigma_g^+) \rightarrow BiO\ (^2\Pi_{1/2}) + CO\ (^1\Sigma^+) \qquad (1)$$

is endoergic (9.4 kcal/mole) and spin allowed, but the reactants and products are predicted[2] by (J, Ω) coupling arguments to be not correlated. These data (and that of Bevan and Husain) show that quenching of Bi $(^2D^o_{3/2})$ state by CO_2 is indeed inefficient. Moreover, the reaction

$$Bi\ (^2D^o_{5/2}) + CO_2\ (^1\Sigma_g^+) \rightarrow BiO\ (^2\Pi_{1/2}) + CO\ (^1\Sigma^+), \qquad (2)$$

also not correlated, is exoergic (2.1 kcal/mole) and spin allowed. Again, the room temperature results show inefficient quenching, but here further

support for the model comes from the observed activation energy, which indicates the exoergic reaction channel is not readily available.

By way of contrast, however, consider the following processes:

$$Bi\ (^2D^o_{3/2}) + O_2\ (^3\Sigma^-_g) \rightarrow BiO\ (^2\Pi_{1/2}) + O\ (^3P_J) \qquad (3)$$

$$Bi\ (^2D^o_{5/2}) + O_2\ (^3\Sigma^-_g) \rightarrow BiO\ (^2\Pi_{1/2}) + O\ (^3P_J) \qquad (4)$$

Here, reaction (3) is spin allowed and slightly endoergic whereas reaction (4) is exoergic by about 11 kcal/mole. Both of these processes are predicted to involve non-adiabatic transitions in that there are no thermally accessible correlations between reactants and products; however, the temperature dependent data indicates an activation energy for reaction (3) of order the endothermicity, and no barrier whatsoever for reaction (4), suggesting an extremely efficient relaxation process is occurring. By utilizing the correlation diagram for these processes, one would not have anticipated this observed behavior. In fact, the (J, Ω) coupling correlation diagrams suggest[2] that quenching by H_2 (D_2), CO, N_2, as well as CO_2 and O_2 (discussed above) must occur via non-adiabatic transitions. The problem in interpreting these data might then be construed as the ability to anticipate when non-adiabatic processes might be efficient and thereby dominate the relaxation mechanism.

One sterling example of a non-adiabatic relaxation mechanism involves likely E→V energy transfer processes. The importance of resonance effects has been discussed earlier[11] and demonstrated by a number of workers[6, 7]. It would appear from these data that E→V processes are likely for the quenching of Bi ($^2D^o_{5/2}$) by hydrogen, where a large isotope effect is observed, viz.

$$Bi\ (^2D^o_{5/2}) + H_2 + 41\ cm^{-1} \rightarrow Bi\ (^2D^o_{3/2}) + H_2\ (v = 1) \quad (5)$$

$$Bi\ (^2D^o_{5/2}) + D_2 - 1026\ cm^{-1} \rightarrow Bi\ (^2D^o_{3/2}) + D_2\ (v = 1) \quad (6)$$

The observed large rate constant for reaction (5) can thus be explained by the possibility of near resonant E → V energy transfer. In addition, these observed temperature dependent results for bismuth can be compared to similar mechanisms suggested[8] for the relaxation process:

$$Pb\ (^3P_2) + D_2 + 163\ cm^{-1} \rightarrow Pb\ (^3P_1) + D_2\ (v = 1)$$

A comparison of the Arrhenius parameters is provided in Table III. The Table lists the A factors, the measured activation energies, and the reaction endothermicity calculated for an assumed E → V process exciting the hydrogen or deuterium into v = 1 with $\Delta J = 0$.

The relative ease by which the quenching of the $^2D^o_{5/2}$ state by H_2 can be rationalized by invoking an E → V transition is to be contrasted by the observed quenching for both states by CO and the $^2D^o_{5/2}$ state by N_2. These molecules would be required to quench these bismuth states via non-adiabatic transitions according to the correlation diagrams constructed using (J, Ω) couplying approximations;[2] both are observed, however, to quench these states showing reasonably large Arrhenius A factors and very large activation energies (see Table II). Any E → V processes relaxing the $^2D^o_{5/2}$ state would be quite far off-resonance and involve $\Delta v = 2$ transitions. In fact, it would be difficult to fully explain these observed rate parameters with any credibility. One mechanism, which may provide some rationalization, involves relaxation due to thermally accessible curve or surface crossings perhaps taking place at small internuclear separations.[6, 11] In this model, the

TABLE III

ARRHENIUS PARAMETERS FOR BISMUTH AND LEAD

	BISMUTH ($^2D_{5/2}$)	LEAD ($^3P_2$)
Q	H_2	D_2
A (cm^3/sec)	4.7×10^{-11}	2.9×10^{-11}
E_A (kcal/mole)	0.9	0.8
ΔE (kcal/mole)	0.4	0.5

observed activation energies reflect the energy of the curve merging complex measured up from the energy of the collision partners at infinite separation. Similar consideration has been postulated to explain the observed large activation energies for cases where there are available exoergic adiabatic routes, such as in the Sn + N_2O kinetics.[12]

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